$\delta^{13}C$ values in archaeological ^{14}C-AMS dated charcoals: assessing mid-Holocene climate fluctuations and human response from a high-resolution isotope record (Arslantepe, Turkey)

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Abstract

RATIONALE: Past climate has always influenced human adaptation to the environment. In order to reconstruct palaeoclimate fluctuations and their role in the evolution of Near Eastern societies during the mid-Holocene, high-resolution $\Delta^{13}C$ records from fossil wood remains at the archaeological site of Arslantepe (eastern Turkey) have been developed.

METHODS: After chemical treatment, $\delta^{13}C$ values were measured by sample combustion flow using a FLASH EA-CHNS instrument interfaced with a Delta V isotope ratio mass spectrometer via a CONFLO III. Two replicates per sample were analysed. The measurement precision was evaluated by propagating variations of the $\delta^{13}C$ values of samples and V-PDB standards, whereas the accuracy was checked by a quality control sample. To account for changes in atmospheric CO_2, $\Delta^{13}C$ values were calculated. In addition, $^{14}C/^{12}C$ ratios were measured by means of ann AMS system (3 MV tandem accelerator).

RESULTS: Mean $\Delta^{13}C$ curves of deciduous *Quercus* and *Juniperus* from archaeological levels between 4700 and 2000 BC (Arslantepe periods VIII-VI D) were produced, where the isotope values were ordered by the available RC ages. Interspecific variations of evergreen vs deciduous plants were postulated for the juniper $\Delta^{13}C$ values being higher than 3‰. The seasonal rainfall amount was recorded by the juniper remains, while the water table levels were obtained from the oak samples.

CONCLUSIONS: The local climate experienced times of enhanced/reduced precipitation in concert with regional trends. Anomalies in the air mass circulation from the Mediterranean basin also produced oscillations of rainfall amount. In such a frame the Rapid Climate Change dry events had a consistent signature in the Arslantepe $\Delta^{13}C$ record, thus potentially contributing to social or organisational changes at the site.

Key words: Palaeoclimate, Mid-Holocene, Charcoal, Stable carbon isotopes, Radiocarbon dating

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1 INTRODUCTION

The Mediterranean region is considered highly sensitive to climate change.[1] As a current research topic, the effects of climate fluctuations on the development of ancient societies have been highlighted by an array of palaeoclimatic proxies.[2, 3] In particular, the semi-arid regions of the eastern Mediterranean, where early state centres arose, are suitable for exploring critical interactions between climate, environment and complex societies.[4, 5] All ages are indicated as years before Christ (BC), even if age scales in years before present (BP) are also shown in the Figures. Climatic, environmental and cultural reasons have all been invoked to explain the increase in social complexity from 5000 BC.[6] Nonetheless the roles of nature- or human-forced changes at the local scale are difficult to disentangle in the absence of on-site and/or high-resolution palaeoenvironmental data.[7]

The demand for mid-Holocene climate reconstruction from Near Eastern archaeological sites has steadily increased over times and several proxy records have been built.[8] First, plant assemblages have been used for palaeoenvironmental reconstruction. In particular, archaeobotanical analyses have focused on anthracological studies since charcoals are available in large amounts from archaeological contexts.[9] In fact in semi-arid environments the most significant factor limiting wood growth is precipitation. Charcoal assemblages though may reflect also human selection of some species, rather than solely humidity and vegetation changes.[10] Secondly, regional pollen,[11-13] lake sediment[14] and speleothem[15] records have provided a useful framework for palaeoenvironmental reconstructions. However, information on spatial variability for past climate trends is still demanded.

Within such a framework, the recent use of stable carbon isotope analysis on archaeological wood remains provides a direct approach to solve this need. Indeed the physiological relation between stable carbon isotopes in fossil plants and atmospheric humidity has widely been used as a tool for the inference on the “free-from-human-influence” palaeoclimate.[16] In recent years several researchers have helped to clarify topics such as precipitation regime,[17, 18] rainfall seasonality,[19, 20] irrigation practices,[21] land use and the impact of human activities.[22]

As a methodological issue, the stable carbon isotope composition ($\delta^{13}C$ value) in C3 plants is influenced by several processes.[23] During the atmospheric CO_2 fixation through photosynthesis, different isotope fractionations occur: (1) the first step takes place during the air collection when stomata opening affects the CO_2 concentration and assimilation rate in the leaves; (2) another process is tied to the activity of the RuBisCO enzyme during the photosynthesis.[24] If water stress occurs, plants close their stomata preventing water loss by leaf transpiration. The abrupt reduction of stomata conductance causes a decrease in the CO_2 concentration within leaf intercellular spaces. Consequently, ^{13}C discrimination also decreases and the $\delta^{13}C$ values of synthesis organic compounds increase.[24] These external and internal factors contributing to the isotopic signature in plant tissues are also affected by other environmental variables such as temperature, solar radiation, relative humidity, vapour pressure deficit and soil moisture.[25-27] Nevertheless it is the physiological interaction between the concentration of atmospheric CO_2 and water availability which mainly influences the $\delta^{13}C$ values of wood.[28]

The archaeological site of Arslantepe is an outstanding long-term settlement in the modern Malatya region (south-eastern Turkey). It provided evidence of one of the first state systems in the Near East dated back to 3400 BC.[29] Huge amounts of charred plant remains were analysed, accounting for land use in the region during the 4300-2000 BC interval, namely in the Late Chalcolithic (hence LC) and Early Bronze Age (hence EBA).[10] In very recent years, stable isotope records at local scale have been established in order to describe climatic conditions behind social transformations at the site. Deciduous *Quercus* (oak) and *Juniperus* (juniper) $\delta^{13}C$ values from 3200-2000 BC have been analysed in order to investigate the response of plant taxa to climate variability.[18, 20] Moreover, C and N stable isotope analyses have been applied on cereal grains assessing the irrigation footprint of crops from 4700 to 2000 BC.[30, 31] Physical models were also compared with charcoal isotope data, enhancing the palaeoclimate reconstruction for the study area.[32]

The present study aims first to extend the time interval of the stable carbon isotope analysis by including charcoal remains from the earliest Arslantepe occupational periods. Secondly, it aims to improve the chronological framework through new ^{14}C-AMS dating carried out on the same plant remains. As a whole, we achieved the first high-resolution isotope records from an archaeological site for the period 4700-2000 BC, displaying more than 2500 years of uninterrupted climate changes and tracing back to the first steps of early state, possibly led by climate constraints.

2 MATERIALS AND METHODS

2.1 Environmental background

The multi-period settlement of Arslantepe is located at the edge of the alluvial Malatya plain along the Euphrates river in south-eastern Turkey (38° 22' 55"N, 38° 21' 40"E, 940 m a.s.l.; Figure 1A). The Anti-Taurus Mountains enclose the plain having the highest peak in the southern Doğanşehir district (2660 m a.s.l.). The regional climate is marked by extreme continental conditions that are mediated to varying degrees by different water resources and geomorphological heterogeneity (Figure 1B). Moisture mainly derives from the Atlantic and the Mediterranean basins.[33] At present, the plain has an annual mean precipitation of 400 mm/yr and a mean monthly temperature between -1 °C and +22 °C.[18] Rainfall occurs mainly in April-May followed by the driest and hottest months of July-October[34] (Figure 1C). The plain is rich in water, with many streams and springs.[35] The hydrogeological catchment is recharged by precipitation on the Anti-Taurus, that amounts to 600/1000 mm/yr and is mostly in the form of winter/early spring snow.[36]

Altitude strongly influences vegetation cover in the region.[37] *Pinus sylvestris* L. (Scots pine) forest is widespread in cold and sub-humid conditions between 2000 and 2700 m a.s.l., together with mountain steppe and grassland vegetation.[38] In addition, *Fagus orientalis* Lipsky (beech), *Alnus* spp. (alder), *Castanea sativa* Mill. (chestnut) and *Cedrus libani* A.Rich. (cedar), *Abies cilicica* Antoine & Kotschy (Taurus fir) occur in broadleaved and conifer forest, respectively, on the belt up to 2000 m a.s.l.[38] The semi-open forest of deciduous broadleaved trees (deciduous and semideciduous *Quercus*; Rosaceae with many species of *Crataegus* L.) and conifers (*Pinus brutia* Ten., red pine; *Pinus nigra* J.F. Arnold, black pine), spaced out by *Juniperus* spp. maquis, dominates from 1200 to 2000 m a.s.l.[38] The semi-arid climate leads to the presence of shrub-steppe vegetation in the valleys and lowlands. However, the particular hydrogeological features of the Malatya plain favour riverine gallery forest, with water-demanding trees, such as *Populus* spp. (poplar), *Salix* spp. (willow) and *Alnus* spp. (alder). Alluvial sediments also provide fertile soils for crop production as in the past, influencing modern vegetation which is mainly characterised by apricot orchards.[39, 40]

2.2 Site setting

The archaeological site of Arslantepe has been excavated by the Italian Archaeological Expedition in Eastern Anatolia, of the Sapienza University of Rome, since 1961. Chronologically, the site can be placed as early as 4700 BC, wto which date the first settlement so far extensively investigated has been dated (see section 3.1).[41] The occupation of this site is exceptionally long and continued until historical times. A recent paper of Vignola et al[31] has reviewed and improved the chronology of the site that has been modelled through the interpolation of the sequence of archaeological levels and 98 radiocarbon dates (Table 1). Specifically, the present work focuses on the contiguous levels from 4700 to 2000 BC. During this timespan, the site gradually developed from rural settlements of LC 1-2 (Arslantepe period VIII, 4700-3900 BC) to the religious and elite centre of LC 3-4 (period VII, 3900-3400 BC) up to the early state centre of LC 5 (period VI A, 3400-3100 BC).[42] At ca 3100 BC the VI A palace, built by the gradual addition of administrative and religious structures and private residences, was destroyed by an extensive fire that ended the centralised system. During EBA I an alternating dominance of different communities occurred in the Malatya plain, with herders' short-lived occupations and a farmers' village (period VI B1 and VI B2, 3100-2750 BC).[42, 43] After a short abandonment but a remarkable cultural break, new short- and long-term settlements succeeded during EBA II-III (period VI C, 2750-2500 BC and VI D, 2500-2000 BC).[42]

The archaeobotanical research at Arslantepe has been carried out for more than 35 years. Several studies have focussed on charcoal remains.[10, 44-46] Architectural woods were abundant since roof structures, and sometimes walls, were made of beams and smaller sticks. Moreover, some of the buildings were supplied with hearths, ovens and other domestic features, in which wood remains were preserved by charring. Archaeobotanical analysis especially revealed that the wood resources were exploited from two main vegetation zones: the plain, where deciduous oaks probably grew, and the surrounding mountain and hill slopes, where juniper trees were available.[10]

2.3 Archaeobotanical samples

A total of 34 charcoal samples, 23 of deciduous *Quercus* and 11 of *Juniperus* sp., were recovered and analysed for this study, joining with 123 samples previously reported by Masi et al.[18, 20] They were mainly selected from the 4700-3400 BC levels (VIII and VII periods) in order to extend back in time the previously available stable carbon isotope records to the earliest LC 1-4 occupation. In addition, charcoals from new deposits of 3400-3100 BC levels (VI A and VI B1 periods) were recovered and selected during the 2012-2015 field campaigns.

Each sample comes from one well-sealed contextual unit, represented by a bin, a fire installation or a wooden structure. The selected samples consist in fragments of young (only few-years old) branches or small sticks. Huge beams from ceilings/roofs, with a high number of tree rings, were avoided since the stable carbon isotope ratio refers to the $\delta^{13}C$ values of atmospheric CO_2 at the time of plant growth.[24] These could have been misleading since the architectural woods might have been in use for a long period of time, thus far later than the time of growth of the tree.[47] Each fragment was carefully cleaned and the last tree-ring was separated using a scalpel. The whole tree-rings were submitted for carbon isotope analysis since several studies point to the preservation of palaeoclimatic signal in the whole tree-ring rather than solely in the latewood, in both soft- and hardwood species.[48, 49]

In order to obtain high-resolution isotope records, charcoals from dated contexts have solely been preferred. For this reason, ^{14}C-AMS dating was additionally performed on 4 selected plant remains. With regard to early period VIII, the remains of a juniper tree branch was selected. In addition, fragments of one juniper branch and two small oak woods were recovered from VI A and VI B1 contextual units, respectively. In these cases, the charcoal fragments were submitted for both ^{14}C and stable isotope analyses.

2.4 Chemical treatment

Radiocarbon (RC) and stable carbon analyses were performed at the CIRCE Laboratory (University of Campania "Luigi Vanvitelli", Caserta, Italy). Non-structural materials were removed from the selected specimens by a Acid-Alkali-Acid (AAA) treatment.[50] Samples were soaked in 3% HCl and 3.2% NaOH for 1 h alternately at 80°C, ending with an additional acid attack (to remove the CO_2 absorbed during the alkali attack) and rinsing the charcoals repeatedly with distilled water. In order to prevent the loss of material, especially during the alkali and demineralised water steps, each fragment was kept in hot-sealed teflon envelopes. Nevertheless in some cases the chemical treatment prevented us from having enough material for the analysis. All the samples were then oven-dried at 70°C overnight before being milled to a powder for isotope analyses.

2.5 Radiocarbon dating

In order to obtain RC determinations, 4 selected AAA-treated samples were combusted and graphitized[51] and the $^{14}C/^{12}C$ ratios were measured by means of an AMS system based on the 3 MV tandem accelerator pelletron 9SDH-2. A detailed description of the ultrasensitive accelerator system of the CIRCE Laboratory is reported in Terrasi et al.[52] The ^{14}C abundances were then converted to RC ages.[53] Calibrations were carried out through CALIB7[54] using the INTCAL13.14c data set.[55]

2.6 Stable isotope and statistical analyses

As reported by Hall et al[56], the climatic signal recorded by $\delta^{13}C$ values in wood tissues is not affected by the carbonization process between 100 and 475°C. Moreover during the charcoalification process biomolecules (cellulose, hemicellulose and lignin) disappear, changing into a new chemically C-enriched material. For this reason no intrinsic differences in the chemical composition of charcoal are expected and the whole wood can be suitable for isotope analyses.[57] In addition, diagenetic degradation of the graphite-like phase in fossil charcoal is not significant and oxidation products can be removed through chemical treatment.[58]
0.30 mg of charcoal was weighed into tin capsules and combusted using a FLASH EA-CHNS 1112 elemental analyser (Thermo Fisher, Bremen, Germany) interfaced with a Delta V isotope ratio mass spectrometer (Thermo Fisher) via a CONFLO III (Thermo Fisher). The isotopic composition is expressed in the $\delta^{13}C$ notation relative to V-PDB (Vienna-Pee Dee Belemnite):

$$\delta^{13}C = \frac{^{13}R_{sample}}{^{13}R_{V-PDB}} - 1$$

where ^{13}R is the $^{13}C/^{12}C$ ratio.[59]
Two replicates per sample were analysed. The standard deviation (SD) of replicate measurements averaged 0.1‰. The accuracy of measurements as the SD of the $\delta^{13}C$ values of the quality check sample (tobacco leaves) was 0.3‰ after sample raw $\delta^{13}C$ normalization and linearity offset error propagation during normalization.
In order to compare plant remains from different archaeological periods, all changes in $\delta^{13}C$ values of atmospheric CO_2 during the Holocene have to be taken into account.[60] For this reason, the carbon isotope discrimination ($\Delta^{13}C$) was calculated as follows:

$$\Delta^{13}C = \frac{\delta^{13}C_{air} - \delta^{13}C_{plant}}{1+\delta^{13}C_{air}}$$

where $\delta^{13}C_{air}$ and $\delta^{13}C_{plant}$ represent the atmospheric and plant $\delta^{13}C$, values respectively (adapted from Farquhar et al[61]).
Past $\delta^{13}C_{air}$ values were inferred by interpolating data from ice-core records[62, 63] and the CU-INSTAAR/NOAA-CMDL database of modern stations.[64] According to the available $\delta^{13}C$ curve of atmospheric CO_2 (http://web.udl.es/usuaris/x3845331/AIRCO2_LOESS.xls), the values applied to the Arslantepe charred woods ranged between −6.29‰ (4700 BC) and −6.40‰ (2000 BC).
Differences in the $\Delta^{13}C$ values of the selected taxa in relation to the archaeological periods were tested through a permutational multivariate analysis of variance (PERMANOVA), significant if $P < 0.05$ (PAST 2.0).

3 RESULTS

3.1 Improved chronological framework

The results of 4 ^{14}C-AMS dates on the selected charcoals are reported in Table 2. They have, together with other recent AMS ages from charred cereal grains, been published and discussed by Vignola et al[31], and have allowed us to distinguish phases within the site's periods. The improved chronological framework, based on old RC ages, stratigraphic sequence and new AMS dates,[41] was used to develop the high-resolution isotope records discussed further in this work.
Looking at the RC ages here presented, the first phase (LC 1) of the earliest occupation at Arslantepe (period VIII) has been dated for the first time.[65-67] The RC age of sample D-DSH7173, ranging from 4720 to 4560 BC (2σ), proves that the LC 1 settlement was established before this date, as already suggested by comparisons with archaeological evidences of other Near Eastern sites.[68]
Despite its recovery in the VI A palace, the calibration range of sample S05/17-DSH7982 from 3639 to 3386 BC (2σ) reveals that this charcoal is archaeologically pertaining to the late-VII period. Indeed it was part of an older furniture, probably a wooden throne, re-used during the LC 5.[69] This attribution was applied to chronologically order the sample within the juniper isotope record.
Finally, significantly the two deciduous *Quercus* RC ages entail continuous short-term occupations at the site after the destruction of the LC 5 settlement (the VI A palace was destroyed around 3100 BC). In fact three phases within the succeeding VI B1 period have been identified, spanning between 3100 and 3000 BC. Such a new chronology of Arslantepe, moreover, has produced new data on the early beginning of the EBA I in the Near East.[41]

3.2 Carbon isotope composition

The $\delta^{13}C$ values with SD of each analysed sample of deciduous *Quercus* and *Juniperus* sp. are displayed in Figure 2, whereas the mean $\delta^{13}C$ values with standard errors grouped by archaeological periods and phases are reported in Table 3.
The $\delta^{13}C$ values of oak samples from three contemporary contextual units of the early-VIII period average at –24.7 ± 0.4‰, and during the late-VIII period the $\delta^{13}C$ value is –24.2 ± 0.2‰. In the early-VII period the mean $\delta^{13}C$ value is established at –25.0 ± 0.2‰. From 4 selected contextual units of the late-VII period the $\delta^{13}C$ values average –24.5 ± 0.2‰. The mean $\delta^{13}C$ value displayed by 19 samples of the VI A period is –24.3 ± 0.2‰. At the

beginning of the VI B1 period $\delta^{13}C$ value is –25.5 ± 0.3‰, but during the mid-VI B1 the selected 19 charcoal samples display a mean $\delta^{13}C$ value of –24.6 ± 0.2‰. At the end of VI B1 the mean $\delta^{13}C$ value from the small oak remains is established at –24.2 ± 0.4‰ on average. The statistical analysis shows that the oak samples from all the investigated periods are similar to each other (Table 3).
The juniper samples, from the early-VIII period charcoals, display a mean $\delta^{13}C$ value of –22.4 ± 0.5‰ whereas at the end of the period the $\delta^{13}C$ value is established at –23.2 ± 0.2‰. Different wood fragments from the same unit of the early period VII record a mean value of –22.2 ± 0.3‰ and at the same time the $\delta^{13}C$ value of the sample of late-VII is –22.1 ± 0.0‰. Finally, the $\delta^{13}C$ values from 13 fragments of juniper wood recovered in contextual units of the VI A period average at –22.2 ± 0.1‰. As reported in Table 3, statistical analysis shows that the VI C values significantly differ from those of all the other archaeological periods.

4 DISCUSSION

4.1 Differences in deciduous *Quercus* and *Juniperus* $\delta^{13}C$ values

The first points to discuss are tied to the variation of the stable carbon isotope ratios in different woody plants and the different ecological behaviours of an evergreen conifer such as juniper and of a mesophilous tree such as deciduous oaks.
The intraspecific variation of $\delta^{13}C$ values in all the site periods ranges from 0.4 to 3.7‰ for deciduous oak and from 0.6 to 1.7‰ for juniper remains (Table 3 and Figure 2). These values largely fit for the intra-population variability that is expected for plants of the same species growing at the same site (over 1.5‰ in arid regions).[70] Nonetheless the main differences have been found within deciduous oak charcoal samples since this taxonomic group gathers together remains of both deciduous and semi-deciduous *Quercus* species (a difference between plant species up to 4‰ is reported in the literature).[70] In addition, deciduous oak specimens exhibit a larger offset in $\delta^{13}C$ values from 3400 to 2000 BC (LC 5-EBA III), where samples from several contextual units were collected (Table 3).
Furthermore, the interspecific variation between the Arslantepe deciduous oak and juniper $\delta^{13}C$ values reflects different physiological features for plant growth (deciduous vs evergreen trees). Indeed environmental factors, and above all water availability, differently influence photosynthesis rates, stomata conductance and RuBisCO activity between the two taxa.[71] In particular, the lower conductance of conifers than of deciduous broadleaved trees may cause the higher juniper $\delta^{13}C$ values here presented (Lloyd et al [72] reported a $\delta^{13}C$ value increase of 3‰). The two selected trees do not anyway show parallel $\Delta^{13}C$ curves as might be expected considering the physiology and autoecology of modern plants, and some isotopic effects reported for plants growing in different altitudinal transects and exposures (slope/plain).[18, 27, 73] are not enough to explain the asynchronicity between the two curves (Figure 4).
A possible mechanism explaining the delay in the deciduous oaks $^{13}C/^{12}C$ curve compared with juniper one was hypothesized by Masi et al.[20] In the Arslantepe record, contrasting ratios in juniper and deciduous oaks suggest that, in addition to seasonality in rainfall distribution, aquifer recharge played a complex role as the Malatya plain is in a karst system. Junipers were probably growing on the slopes surrounding the plain where oaks were widespread. Junipers used late winter/early spring rainwater, while deciduous oaks mostly water recharged from the aquifers during late spring and early summer. As the time needed to recharge the water table in a karst area can be estimated at some decades, the water or part of the water that the two species used for photosynthesis had a different age.

Thus, the present study confirms that the Arslantepe juniper record is mainly influenced by local precipitation during rainy seasons, whereas the Arslantepe oak, being tied to summer droughts, registers the water isotopes content of some decades before.
This behaviour in confirmed by the investigation of the co-occurrence patterns of the carbon isotope discrimination in deciduous oak and juniper remains. Samples of the two taxa recovered from the same contextual unit (i.e. the same time period) have been plotted in Figure 3. It is clear that the relationships between $\Delta^{13}C$ values of deciduous oak and juniper from 4700 to 2000 BC are positive and statistically significant only in few cases (r from 0.69 to 1). As confirmed by modern samples ($r = 0.17$), the isotopic content of the two plants must be interpreted in a different way.

4.2 Changes in local precipitation and cultural developments

The second guideline of the discussion is focussed on the palaeoclimate reconstruction in the study area and the comparison with archaeological evidence.
The mean $\Delta^{13}C$ values of the two selected taxa from all the investigated sequences are plotted in Figure 4. The carbon isotope composition of the fossil woods compared with the modern ones indicates that the past climate was generally wetter than the climate recorded in 2008 (350 mm/yr, Figure 1C). This difference, already reported by Masi et al[18, 20] for 3400-2000 BC, is here confirmed as being since around 4700 BC, with the only exception of the period around 2300 BC, when oaks registered more aridity than today.
As stated by Masi and colleagues[20], water sources, precipitation pattern and hydrogeological features at the local scale influenced the isotope composition. Differences in $\Delta^{13}C$ trends of the Arslantepe deciduous oak and juniper, previously reported, have been confirmed here. Significantly, the importance of the dataset and the methodological approach are validated.
Since the growth of deciduous oaks occurs at spring/summer when the local precipitation rate is low, their high rooting-depth supplies them with effective uptake of groundwater in the plain during dry months. In a karst environment such as the Malatya area, the differences in plant auto-ecology and distribution (deciduous oak probably grew in the plain fed by karst aquifers, whereas juniper in the slopes of the surrounding mountains) account for the time lag existing between rainfall amount and aquifer recharge, leading to a quasi-regular shift in the dec. *Quercus* isotope signal (Figure 4).[20]
In the Malatya area water availability resulted therefore in a dependence on 1) the seasonal distribution of rainfall, having its climax between March and June (Figure 1C and 2) on the several karst springs in the plain.
We should therefore suppose that the curve directly linked to rainfall is the juniper one, while the deciduous oaks one shows a delay of some decades due to the time needed for water recharge.
The AAP (Average Annual Precipitation) macrophysical model for the Malatya plain, applied by Arıkan et al[32], displays no overall wetter conditions in the first half of EBA (Figure 4).
From 4640 to 4200 BC, a transition toward wet conditions is indicated by the juniper $\Delta^{13}C$ values. A high moisture level is also testified by isotope data from herbivore bone remains[74] (Figure 4).
Later, from 3700 to 3400 BC, when social complexity gradually grew at the site[29], a decrease in precipitation marked by the juniper $\Delta^{13}C$ values was balanced by the increase in aquifer level reported by the deciduous oak $\Delta^{13}C$ values. The reduced precipitation water is also confirmed by the fact that barley was probably irrigated (we have so far no wheat remains between 4200 and 3200 years BC) (Figure 4).
Soon after 3400 BC both oak and juniper experienced an abrupt change followed by an instability phase established between 3400 and 2900 BC (LC 5-EBA Ia, Figure 4). This

period corresponds to the early state at Arslantepe, which thus certainly had to face such fluctuations. It is noteworthy that the abrupt increase in the deciduous oak $\Delta^{13}C$ values was precisely recorded in the LC 5, when drier and wetter periods have also been pointed out in quick succession by the juniper and hare bone isotope records[74] and the concurrent increase of agricultural pressure on the plain has been reconstructed.[31] Looking at the past crop production in the Malatya area, both barley and wheat fields were always supplied with agricultural practices (such as reserving better fields) when the decrease of rain water is recorded by juniper $\Delta^{13}C$ values[31] as testified by definite thresholds of cereal $\Delta^{13}C$ values as reported by Vignola et al.[31] (Figure 4) A crop production managed by the centralised institutions of Arslantepe[31] would have been able to balance the climate fluctuations in the plain. Notwithstanding this, at the end of the LC 5 the state system lapsed into decline; causes for this are to be sought in long existing to social tensions.[29]

A stability phase with enhanced late winter/early spring precipitation is suggested for the 3000-2500 BC period (EBA Ib-II, Figure 4). Considering the occupational patterns outlined for the Malatya plain during the investigated sequence, the highest juniper $\Delta^{13}C$ values exactly correspond with the establishment of new settlements on the natural hills[75] (Figure 4). Probably, the seasonal variation (= more water available also outside the karst lain) fostered mobility and transhumance of the VI B1 and VI C herder groups.[76] It is noteworthy that the differences between the VI C $\Delta^{13}C$ values and all the other Arslantepe periods are significant (Table 3).

Finally, a consistent dry event is recorded at 2300 BC (EBA III) when the past climate was similar to the present one (Figure 4) and wheat fields were water supplied. This aridity phase, with minimum AAP, has been also highlighted by the stable oxygen isotope ratios from animal remains.[74] This might have been a central factor stimulating the concentration of settlements nearer to the river, into the Euphrates floodplain, as testified by the settlement system of the Malatya plain[75] (Figure 4).

4.3 Regional palaeoclimate fluctuations

The interpretation of the Arslantepe isotope data in comparison with other proxies, aimed at an overall reconstruction of the past regional climate, is the last focus of this study.

During the mid-Holocene the eastern Mediterranean has been sensitive to changes in rainfall and vegetation parameters, while temperature has remained approximately unvaried.[77] In particular, after 5000 BC a transition phase, leading to aridification, emerged.[3, 15] Regarding changes in precipitation, we have to locally consider the juniper isotope record since it has previously been shown that the Arslantepe juniper $\Delta^{13}C$ values are influenced by the precipitation pattern (see sections 6.1 and 6.2). Thus, changes to wetter/dryer conditions are highlighted by the juniper $\Delta^{13}C$ values from 4700 to 1900 BC (Figure 4).

How climate fluctuations have varied in amplitude and duration throughout the history of the Arslantepe site is, however, hard to assess in detail because we have a good, but not always high-resolution record.

The Sofular Cave stalagmite record, reflecting hydrological conditions over the southern Black Sea coast (NW Turkey), shows high rainfall amount, particularly until 4150 BC.[78] This wet period falls within the time frame of warm conditions found in the northern latitudes, referred as 'Holocene climatic optimum' (from ca 7050 to 4050/3050 BC),[79] when the organic-rich sapropel S1 sediments were deposited in the eastern Mediterranean.[80] At Lake Sünnet (W Turkey), after high $\delta^{18}O$ values suggesting hot summers and dry conditions, more humid climatic setting was established from ca 4700 to 4050 BC.[81] In SW Turkey, the $\delta^{18}O$ record from Gölhisar Lake shows a reducing trend until ca 4100 BC, with a precipitation/evaporation ratio considerably higher than at present[82] (Figure 5). Furthermore,

positive variation in the eastern Mediterranean sea-level confirms high rates in the fluvial flow feeding the basin from ca 5000 to 4000 BC (W Turkey and Israel coastline).[83] From the Soreq Cave speleothem (Israel) $\delta^{18}O$ values record the peak of wet climate at ca 4550 BC, when the annual precipitation probably exceeded 700 mm[15] (Figure 5).
From 4150 to 3400 BC a decrease in moisture is recorded in both the Arslantepe area and the Van region (E Turkey), where Wick et al[11] have confirmed the sensitive relationship between relative humidity and the carbonate oxygen-isotope signal from the Lake Van laminated sediments. In particular, at ca 3850 BC the Malatya AAP reached its lowest value (Figure 4). The increase in $\delta^{18}O$ values after 4050 BC in the Gölhisar record has been interpreted as resulting from gradually increasing drier conditions (Figure 5). Considering the contemporary positive shift in the $\delta^{18}O$ signal from Lake Sünnet, combined with the reducing lake level[81], a parallel climate evolution can be envisaged from the south-eastern to western regions. In addition, a long dry period is evident from Jeita Cave, Lebanon.[84] The water-level record from the Dead Sea basin also shows lower depth from around 3800 to 3600 BC with regard to the later sequence, when the lake level grew consistently[85] (Figure 5).
Between 3400 and 3100 BC, when the Arslantepe pristine state rose, oscillations of the regional precipitation pattern occurred. In particular, episodes of greatest isotope variability with large-scale switches in the stable oxygen and carbon isotope ratios appear to represent centennial-scale shifts in the rainfall amount from Soreq Cave and precipitation/evaporation balance at Gölhisar Lake (Figure 5). This suggests marked effects of climate fluctuations on the regional environments. Interestingly, oak charcoal remains from the north Syrian sites display the highest and lowest $\Delta^{13}C$ values during LC 5 in strict accordance with the Arslantepe juniper signals from the same timespan[22] (Figure 5). As in regions not subject to a karst system oaks trend are in agreement with juniper one, this is another evidence of the anomaly of the deciduous oaks curve at Arslantepe. The delay was explained with the time needed for the water recharge.
The high-resolution records from Soreq Cave and Arslantepe clearly indicate climate fluctuations at the end of the LC (3400-3100 BC), not apparent from other low-resolution proxies. This climate instability involved the whole Near East, possibly having a significant impact upon local societies.[8] A drop in humidity at the end of LC/beginning of EBA (3400-3000 BC) is also shown by *Pistacia* pollen and isotope records of Lake Van, eastern Turkey.[11] In the Mediterranean basin, Mayewski et al[86] identified a major arid RCC (Rapid Climate Change) event between 4050 and 3050 BC, with a peak at 3250 BC (Figure 5). Some scholars have already related the dry event of 3250 BC (5.2 ka BP event; ka stands for kiloyears) to a consistent drought-triggered social change in the Late Uruk culture of the Great Mesopotamia.[6] This climate instability could have played a role in exacerbating the crisis of the Arslantepe LC 5 society, as already pointed out.
Despite the Bronze Age aridity trend in the eastern Mediterranean[3], increasing moisture is initially indicated by the Arslantepe, Gölhisar and Sofular isotope records, between 3000 and 2500 BC, together with minor oscillations due to local influences[78] (Figure 5). At Soreq Cave the $\delta^{18}O$ values indicate wetter climate conditions from 2850 to 2750 BC with a mean annual precipitation of ca 700 mm.[15] The elevation of the Dead Sea level also confirms significant water inputs (above 390 m a.s.l. at 3000-2250 BC, Figure 5).
The climate oscillations that occurred at around 2250 BC are differently invoked in the historical reconstruction of the EBA III societies of the Mesopotamian regions.[87-89] The Arslantepe juniper $\Delta^{13}C$ minimum at 2350 BC is framed in the aridity trend evidenced at Eski Acıgöl[90], Lake Van and Soreq Cave oxygen isotope curves (Figure 5). The increased oxygen isotope signal matches with the inflection in the Lake Van oak forest and high micro-charcoal values.[11] Similar forest openings have been highlighted in the pollen records of Mirabad and Zeribar lakes (E Iran) at ca 2200 and 2050 BC, respectively.[91, 92] South of

Turkey, the $\Delta^{13}C$ values from Syrian oaks also reveal drier conditions around 2500-2300 BC. At Soreq Cave a 2200 BC dry event occurred at the end of a long trend towards aridity that started at 2750 BC (Figure 5). Here, although the positive peak of the $\delta^{18}O$ values does not indicate such a severe aridity, the corresponding $\delta^{13}C$ values suggest a significant change in magnitude.[15] This falls within the global-scale RCC event (the so called 4.2 ka BP event)[93] that has a consistent signature in the Arslantepe record, leading to the highest water deficit (Figure 5). Staubwasser and Weiss[6], referring to this event as a 'Collapse as Adaptation to Rapid Climate Change', proposed the relationship between low rainfall and the collapse of the Akkadian Empire. At Arslantepe and in other Near Eastern sites, however, a continuity of the societal developments between the EBA III and MBA periods is now clearly attested.[94] No evidence of a consistent cultural break has been highlighted in the site in the Malatya plain at the end of the EBA III period.[95] The tracking of more favourable habitats into the Euphrates floodplain of the Malatya area in this period may, however, be regarded as a consequence of the drought displayed by the AAP model at 2250 BC (Figure 4). In addition, Arıkan[96] confirms that human communities used high-risk landforms across the Upper Euphrates basin during the EBA III aridity.

Finally, the last marked increase in precipitation at the site (2150 BC) has been traced out in western (Sofular Cave) and central (Nar Gölü)[97] Turkey as well as in the southern regions (Figure 5). Interestingly, Benito et al[98] report extreme flooding events of the eastern Mediterranean rivers since 2150 BC.

It is worth mentioning that the precipitation pattern in the Malatya region also shows similarities with the western Turkey proxies during the wet phases. On the contrary, the trend of reduced moisture is mainly shared with E Turkey and the southern regions. The eastern Mediterranean climate, as a result of the interaction between European, Asian and North African patterns, is mainly affected by the Siberian High Pressure System in winter.[1] Thus, a strong exposure to this System favours cyclogenesis and rainfall from western Russia.[99] Nonetheless, Turkey's areas north of the Taurus range can have experienced influences by the NAO (North Atlantic Oscillation) from the western Mediterranean, following preferred tracks eastward as evidenced by several studies.[100] In addition, Roberts et al[101], focusing on the climate signals during historical times, have shown that there is no single mode that accounts for the association of Atlantic-Mediterranean fronts in the precipitation regimes. For this reason we propose that the changes in rainfall at Arslantepe might have been influenced by the oscillations of the atmospheric circulation in the Mediterranean basin, even if driven by the Atlantic circulation. While the wettest phases could have been tied to anomalies of the North Atlantic Oscillation (NAO) forcing[102], the driest ones could have been caused by the high-pressure system from the eastern regions. Looking at the *Pistacia* pollen record from Lake Van, the increases in percentage of the pistachio pollen grains mostly corresponds with the events of reduced moisture at Arslantepe (Figure 5). On the other hand, when the wettest conditions are recorded in the Malatya area from 3000 to 2500 BC, depletion of the pistachio shrubland is evident around the lake (Figure 5). Since pistachio shrubs are drought-tolerant but frost-sensitive, changes of the precipitation pattern with increasing late winter snow potentially suggested by the Arslantepe juniper record is confirmed.

The aquifer recharge pattern in the Malatya plain, shown in the possible delay in the Arslantepe deciduous oak $\Delta^{13}C$ values, interestingly matches with other palaeoenvironmental proxies. In particular, the carbon isotope record from Lake Hazar (Elaziğ region) close to the study area reveals resemblance in recording palaeoclimate variability (Figure 5). This correlation is probably due to the presence of the karst phenomenon also in that site of the Anti-Taurus region.[103] In addition, fluvial sedimentation phases from neighbouring areas can be compared with the climate changes in the Upper Euphrates valley.[104] In particular, in the Middle Euphrates region the formation of a fluvial terrace, stabilized by dense vegetation, marked the mid-Holocene until 4000 BC, when incision occurred.[105] It is also noteworthy that the low groundwater levels in the Malatya plain correspond with sediment deposition activities of the Upper Tigris valley, suggesting dry phases. On the contrary, the precipitation increase caused the incision on the riverbed and possibly the growth of the local water table[106] (Figure 5). In fact, during wet winters the intra-seasonal soil moisture loss is reduced, fostering effective river flows and discharge into the aquifers.[102]

CONCLUSIONS

It is clear from the present work that interdisciplinary research can help in identifying and understanding how palaeoclimate has influenced social response to palaeoenvironmental changes. Stable carbon isotope analysis on archaeobotanical wood remains from the site of Arslantepe (Malatya, Turkey), where one of the earliest state systems appeared, has successfully led to climate reconstruction during the mid-Holocene. In particular, we have developed $\Delta^{13}C$ records from two plant taxa, deciduous *Quercus* and *Juniperus*, from 4700 to 2000 BC. High-resolution chronology has been obtained by joining the available chronological framing, improved with new ^{14}C-AMS dates, and the outstanding archaeological sequence.

The Malatya region experienced times of enhanced/reduced precipitation in concert with regional trends, but anomalies in the Mediterranean atmospheric circulation were also involved in the local climate oscillations.

From 4700 to 4200 BC wet conditions have been recorded, followed by a slight reduction of moisture. At that time an increasing social complexity was gradually testified at the site. From 3400 an instability phase occurred until 3000 BC, with possible important influence in the evolution of the LC 5 pristine state. We have suggested in particular that the crisis of the Arslantepe centralised organization could have been heightened by the 3250 BC (5.2 ka BP) peak of RCC arid event in the Mediterranean basin, resulting in the intensified effort of the political and economic leaders to improve the land productivity, thus probably increasing the pressure over the population and thereby exacerbating the crisis.

From 3000 to 2500 BC the increase of the late winter/early spring rainfall coincided with the mobile occupation of pastoral groups and the increased number of the archaeological sites settled on the natural hills across the Malatya plain. Finally, at ca 2300 BC (the so called 4.2 ka event) the RCC dry event of the eastern (and central) Mediterranean, having a high signature in the Arslantepe records, coincided with and possibly had a role in the movement of the local communities into the Euphrates floodplain. This was, however, also related to a new cultural and political change, resulting in the establishment of more sedentary communities living in small towns surrounded by walls, and probably competing each other. There was evidence that the crop production was clearly fostered with irrigation practices/favourable locations in the surrounding plain when the decrease of rainwater has been recorded by the isotopic signal. Finally, a substantial continuity in the cultural development of the EBA III and MBA societies has been documented at Arslantepe and in the Upper Euphrates region, when we see again a strong arid event; not a dramatic crisis, but rather a re-arrangement of the political and economic organization of the local communities is attested there at the end of the 3rd millennium BC.

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Author contributions

The manuscript was conceived and written by C. Vignola with contributions of F. Balossi Restelli, M. Frangipane, F. Marzaioli, A. Masi and L. Sadori. C. Vignola, F. Marzaioli, I. Passariello, M. Rubino and F. Terrasi were responsible for data production. C. Vignola, A. Masi and L. Sadori were responsible for data management and interpretation. Figures were created by C. Vignola.

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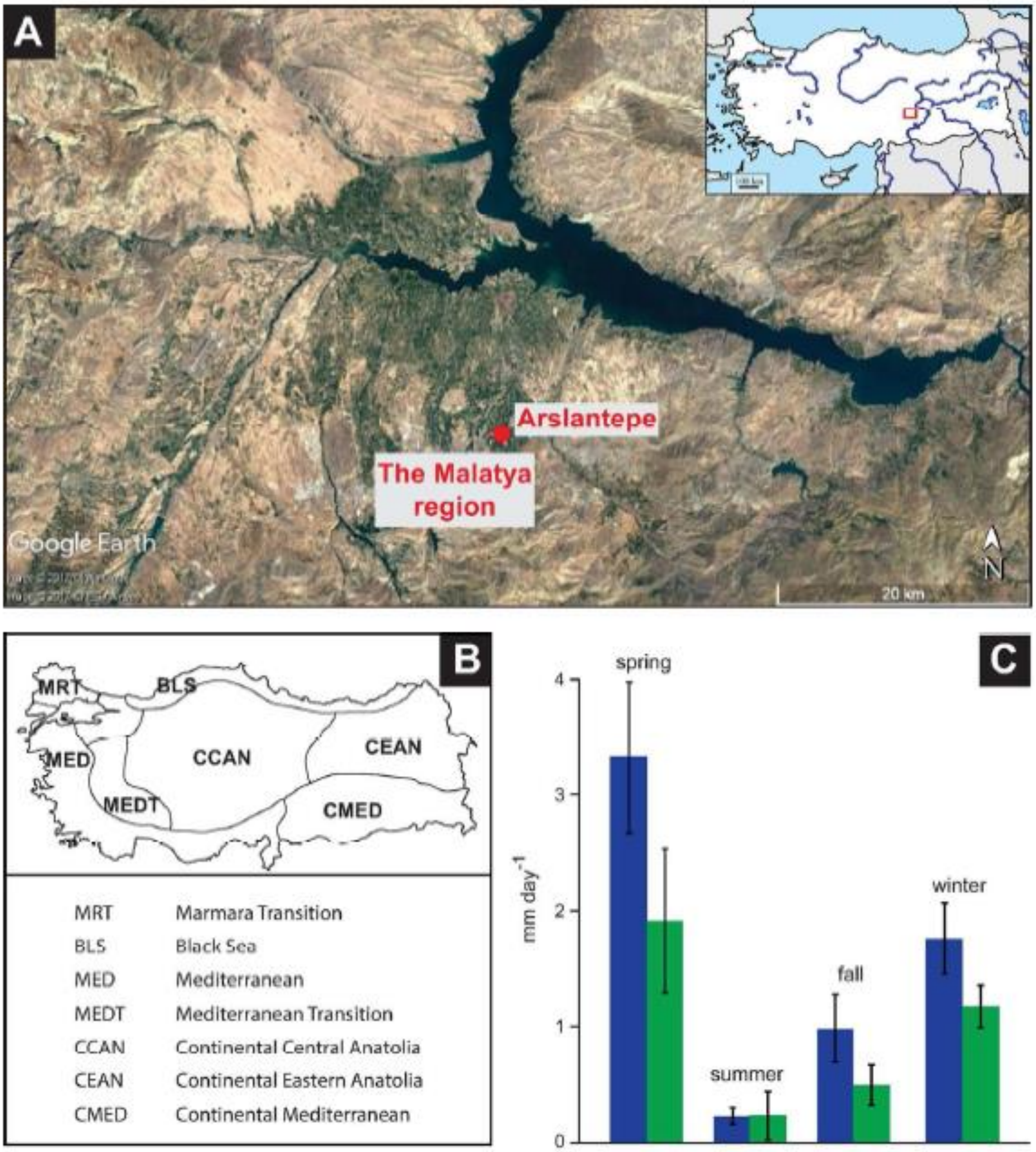

Figure 1 Arslantepe (Malatya, Turkey). A) The Malatya plain with the Arslantepe site (area location top right). B) Modern climate zones of Turkey.[33] C) Seasonal precipitation amounts in the study area: in blue for the period 1961-1990 , in green for 2008.[18]

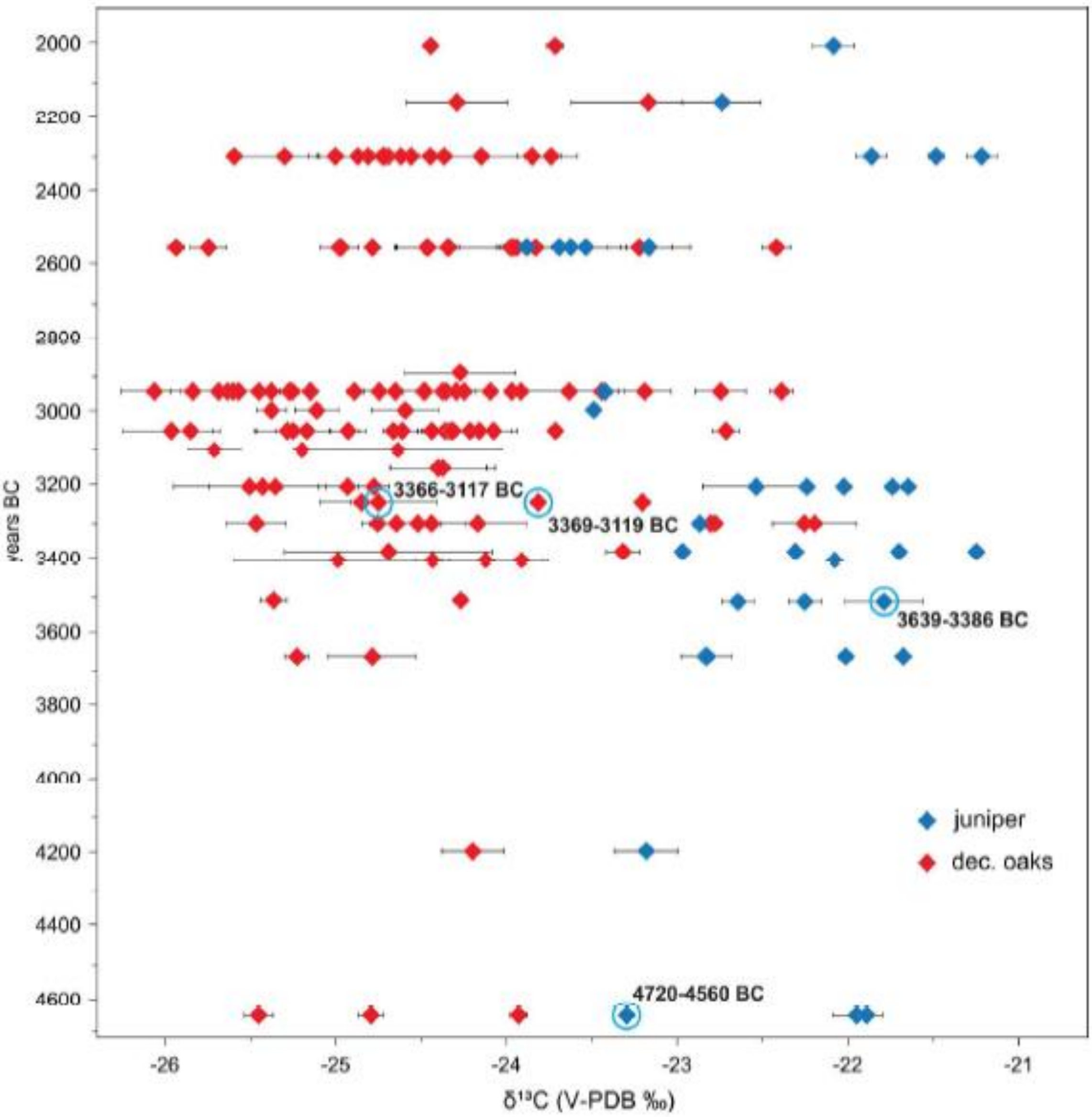

Figure 2 δ^{13}C values of deciduous oak and juniper charcoal samples in the chronological framing. Within circles, samples ^{14}C-AMS dated with their RC ages (Table 2). Error bars for standard deviation (SD).

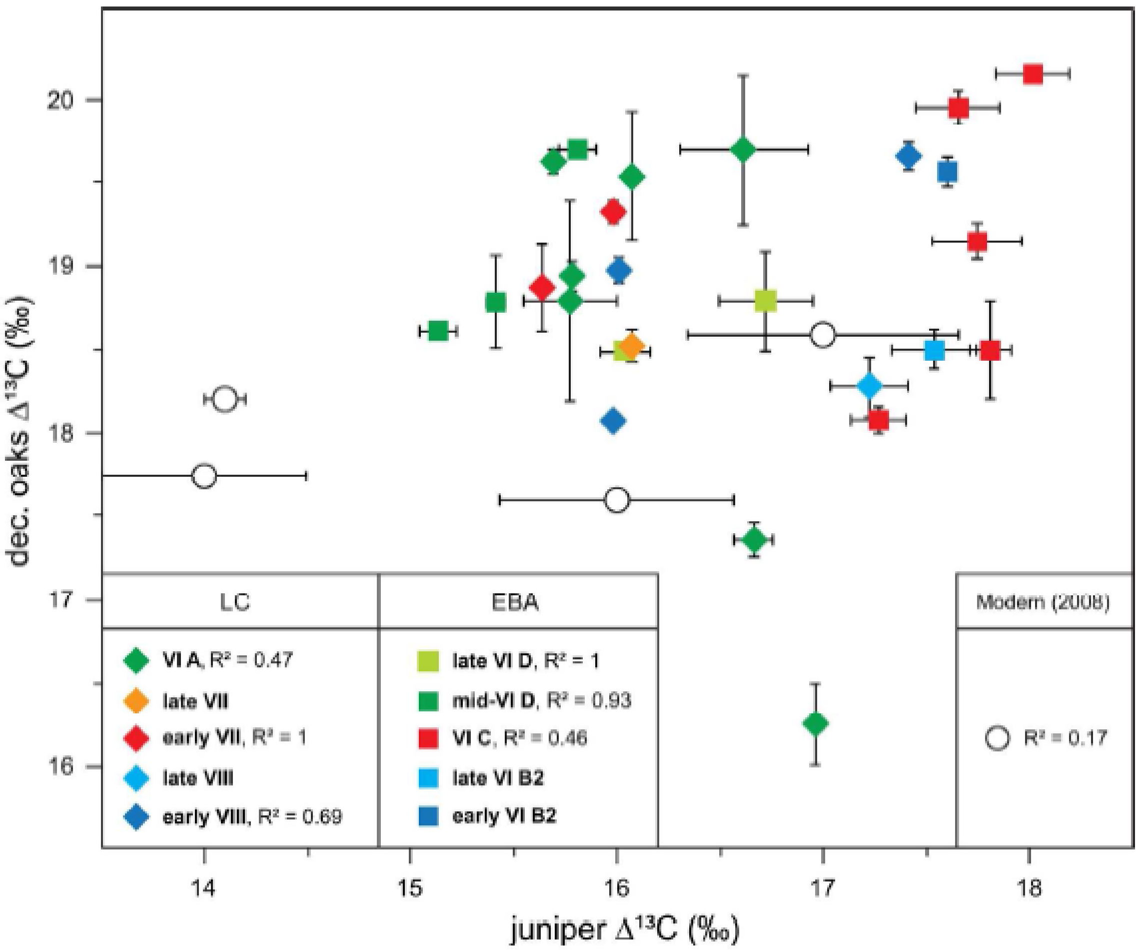

Figure 3 Relationship between deciduous oak and juniper $\Delta^{13}C$ values of charcoal samples from the same contextual unit, grouped by archaeological periods. Error bars for standard deviation (SD).

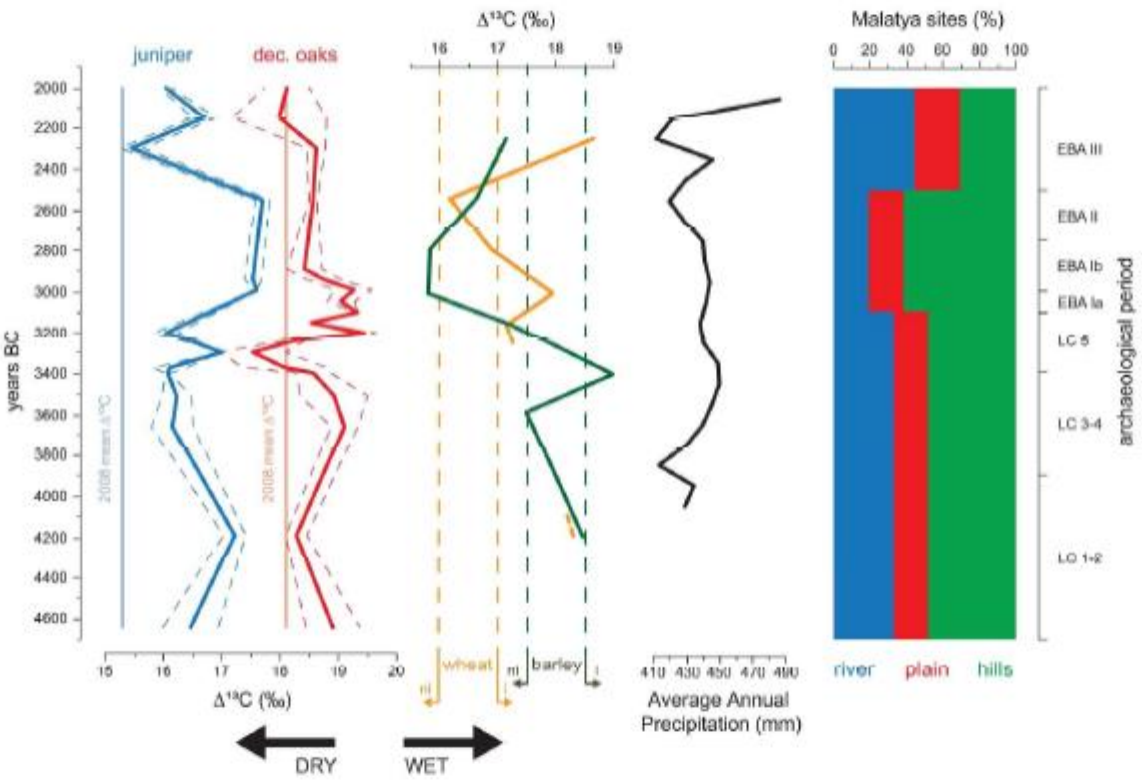

Figure 4 From left to right: $\Delta^{13}C$ mean values with standard errors of deciduous oak and juniper charcoal remains from Arslantepe (vertical dotted lines represent mean isotope values of modern samples; data from this paper and Masi et al.[18, 20]); AAP (Average Annual Precipitation) values from macrophysical model of the Malatya area;[32] $\Delta^{13}C$ mean values of barley and wheat grains from Arslantepe with thresholds for water availability (n.i. for not irrigated, i. for irrigated);[31] % values of site distribution in the Malatya plain.[75]

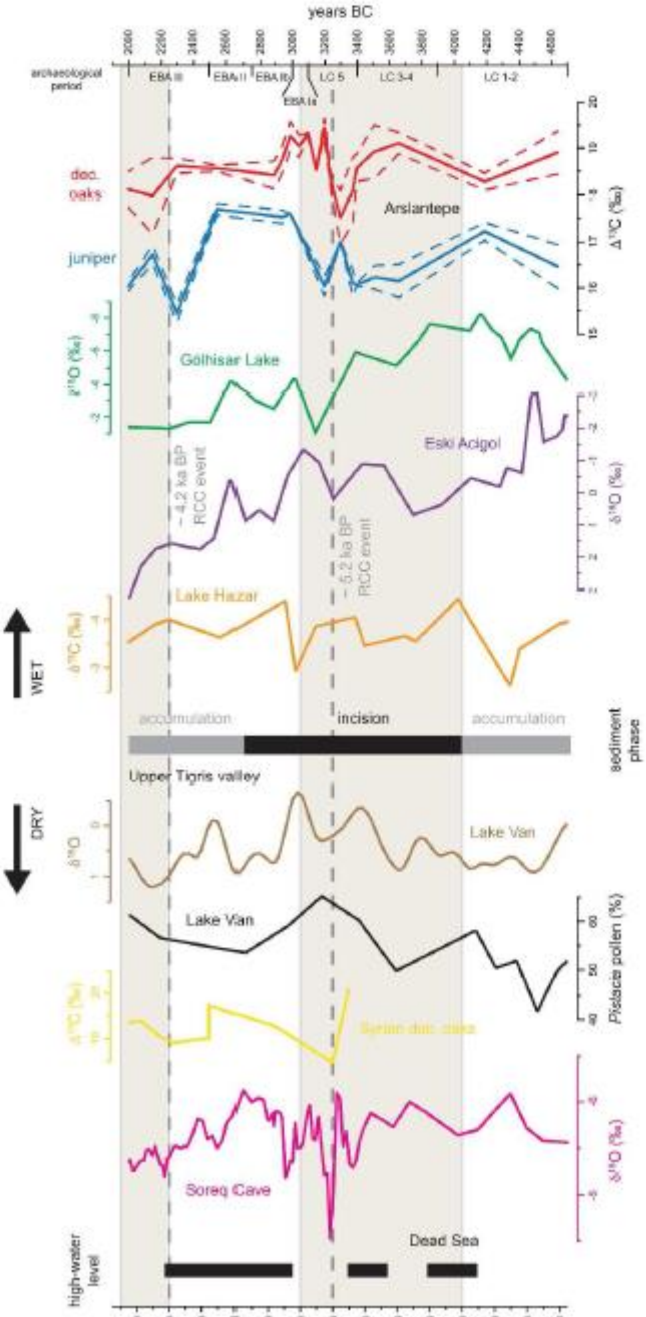

Figure 5 Comparison between Near Eastern palaeoclimate proxies. From top to bottom: $\Delta^{13}C$ mean values with standard errors of Arslantepe deciduous oak and juniper charcoal remains; $\delta^{18}O$ values from Gölhisar Lake;[82] $\delta^{18}O$ values from Eski Acıgöl Lake;[90] $\delta^{13}C$ values from Lake Hazar;[103] fluvial sedimentation phases from Upper Tigris valley;[106] $\delta^{18}O$ and *Pistacia* pollen % values from Lake Van;[11] $\Delta^{13}C$ mean values of deciduous oak charcoals from northern Syria sites;[22] $\delta^{18}O$ values from Soreq Cave speleothem;[15] high-water level of Dead Sea basin.[85] Shaded rectangles and vertical dotted lines indicate the Rapid Climate Changes (RCC dry events) as singled out and described by Mayeski et al.[86]

Table 1 The investigated archaeological periods and the new chronological framing obtained by the interpolation of old Radiocarbon (RC) ages, stratigraphic sequence and new AMS RC dates (see section 3.1).

Chronological sequence of eastern Turkey	Arslantepe period	Years BC
Early Bronze Age III	VI D	2500-2000
Early Bronze Age II	VI C	2750-2500
Early Bronze Age Ib	VI B2	3000-2750
Early Bronze Age Ia	VI B1	3100-3000
Late Chalcolithic 5	VI A	3400-3100
Late Chalcolithic 3-4	VII	3900-3400
Late Chalcolithic 1-2	VIII	4700-3900

Table 1

Table 2 ^{14}C-AMS dates provided on samples from the same contextual units investigated by stable isotope analysis (data from Vignola et al[31] and this paper).

Arslantepe period	Phase	Taxon	Plant remain	Laboratory ID	^{14}C date BP	^{14}C date BC (2σ)
VI B1	Late	deciduous *Quercus*	Charcoal	D28/15-DSH7017	4552 ± 21	3366-3117
	Middle/Late	deciduous *Quercus*	Charcoal	D28/15-DSH7017	4559 ± 21	3369-3119
VI A		*Juniperus* sp.	Charcoal	S05/17-DSH7982	4767 ± 28	3639-3386
VIII	Early	*Juniperus* sp.	Charcoal	D-DSH7173	5801 ± 21	4720-4560

Table 2

Table 3 $\delta^{13}C$ and $\Delta^{13}C$ mean values with standard errors of deciduous oak and juniper charcoal remains for the investigated archaeological periods. Light grey characters: data from Masi et al.[18, 20] Lower-case superscript letters: archaeological periods whose differences are not statistically significant (PERMANOVA). *s* = sample variation (max-min values).

		Phase	Sample (*n*)	$\delta^{13}C$ (‰)	*s* of $\delta^{13}C$ (‰)	$\Delta^{13}C$ (‰)
Deciduous *Quercus*	VI D	Late	4	–23.9 ± 0.3	0.7	18.1 ± 0.4[a, b, c, d, e, f, g, h, i, j, k]
		Middle	16	–24.6 ± 0.1	1.9	18.7 ± 0.1[a, b, c, d, e, f, g, h, i, j, k]
	VI C		15	–24.4 ± 0.2	3.5	18.5 ± 0.2[a, b, c, d, e, f, g, h, i, j]
	VI B2	Late	29	–24.6 ± 0.2	3.7	19.2 ± 0.3[a, b, c, d, e, f, g, h, i]
		Early	3	–25.0 ± 0.2	0.8	18.8 ± 0.2[a, b, c, d, e, f, g, h]
	VI B1	Late	4	–24.2 ± 0.4	1.6	18.3 ± 0.2[a, b, c, d, e, f, g]
		Middle	19	–24.6 ± 0.2	3.3	18.7 ± 0.2[a, b, c, d, e, f]
		Early	3	–25.5 ± 0.3	1.1	19.3 ± 0.0[a, b, c, d, e]
	VI A		19	–24.3 ± 0.2	3.3	18.4 ± 0.3[a, b, c, d]
	VII	Late	6	–24.5 ± 0.2	1.5	18.6 ± 0.2[a, b, c]
		Early	2	–25.0 ± 0.2	0.4	19.1 ± 0.2[a, b]
	VIII	Late	1	–24.2 ± 0.2	-	18.3 ± 0.2
		Early	3	–24.7 ± 0.4	1.6	18.9 ± 0.5[a]
Juniperus sp.	VI D	Late	2	–22.4 ± 0.3	0.6	16.4 ± 0.3[a, b, c, d, e]
		Middle	3	–21.5 ± 0.2	0.7	15.5 ± 0.2[a, b, e]
	VI C		5	–23.6 ± 0.1	0.7	17.7 ± 0.1[d]
	VI B2	Late	1	–23.4 ± 0.2	-	17.5 ± 0.2
		Early	1	–23.5 ± 0.0	-	17.6 ± 0.0
	VI A		13	–22.2 ± 0.1	1.7	16.2 ± 0.1[a, b, c]
	VII	Late	1	–22.1 ± 0.0	-	16.1 ± 0.0
		Early	3	–22.2 ± 0.3	1.1	16.2 ± 0.4[a, b]
	VIII	Late	1	–23.2 ± 0.2	-	17.2 ± 0.2
		Early	3	–22.4 ± 0.5	1.4	16.5 ± 0.5[a]

Table 3